

Laminated Films with Isotropic In-Plane Properties

Commercially available polymer films used in structural applications, such as biaxially oriented poly(ethylene terephthalate), exhibit pronounced anisotropy in mechanical properties, thermal expansion, and long-term dimensional stability. Films with more nearly isotropic in-plane properties have been produced by laminating plies of PET film at various angles to one another. In addition, composite films have been made with nearly isotropic properties and with significantly reduced coefficients of expansion compared to those of commercially available polymer films. A laminate with PET faces bonded to a low expansion alloy foil core had the best dimensional stability and the least anisotropy of the films studied.

Introduction

There are a number of structural applications for polymer films in which isotropic in-plane properties are desired. One such application is in flexible disk magnetic recording media. High density magnetic recording on flexible disks requires that the substrate film be as dimensionally stable and as nearly isotropic as possible. At present, flexible magnetic recording media are composed of a biaxially oriented poly(ethylene terephthalate) film substrate which has been coated with magnetic particles in a polymer matrix. Commercially available PET films have an excellent combination of strength, stiffness, toughness, and low cost. In addition, they have relatively good dimensional stability when compared to other commercial films [1]. However, biaxially oriented PET films exhibit significant anisotropy in mechanical properties and coefficient of thermal expansion [2-6]. Also, oriented PET films are subject to shrinkage over long periods of time due to stress relaxation. This long-term shrinkage is also highly anisotropic [3-7].

Several approaches have been considered for producing flexible films with improved dimensional stability and low in-plane anisotropy. A properly annealed PET film should shrink less than an unannealed film over long periods of time at room temperature and annealing of PET films has been studied by several workers [2, 4-7]. In general, annealing produces films with significantly improved long-term dimensional stability but the annealed films remain anisotropic in mechanical properties and thermal

expansion coefficient. Studies of the dimensional stability of high temperature resistant films such as cast polyimide [6], cast poly(parabanic acid) [7], and biaxially oriented poly(ethylene 2,6-naphthalate) [7] have shown that these films are not more stable than PET at temperatures near room temperature when all of the various dimensional instabilities are considered.

Various polymer film laminates can be made with combinations of properties which are not available in single-ply films [8]. Cross-ply lamination is an approach to producing isotropic in-plane properties in films. Two cross-ply films, both of which are two-ply high density polyethylene films, are commercially available [9, 10] and one Canadian patent [11] on the subject of cross-ply films has been issued. Cross-ply PET film laminates are not commercially available and they have not been reported in the literature.

In the work reported here, PET film laminates were produced by bonding plies of unannealed or annealed PET film so as to form more nearly isotropic films. Thus, in two-ply laminates, the plies were oriented at 0 and 90 degrees with respect to one another. In three-ply laminates, the ply orientation was -60, 0, and +60 degrees. Two- or three-ply laminates with cross-ply orientation can be made in continuous lengths, although they contain joints where film edges meet in a ply. The continuous laminates can be coated using conventional coating equip-

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ment. Laminates of more than three plies can be made using a bonding process; but these laminates offer very limited improvement over three-ply films, and they probably are not cost effective.

While lamination of polymer films can be used to produce isotropic properties, films with maximum dimensional stability require some form of reinforcement. If the reinforcement is properly oriented, the films will also have isotropic in-plane properties. A major problem is that the reinforcements which lead to the best dimensional stability tend to be brittle and high in modulus. To be flexible, the film must be very thin, and to be isotropic, the reinforcement must be precisely oriented and uniformly distributed. Woven and nonwoven fabric reinforcements allow good control of fiber orientation and distribution, and some work on fabric-reinforced films is described in this report.

Certain metal foils have low coefficients of thermal expansion and are not subject to hygroscopic expansion of time-dependent dimensional change. Laminates of polymer films and these metal foils can be made with isotropic properties and good dimensional stability. This requires that the thickness of the metal foil used is such that it neutralizes the anisotropy of the polymer film in the laminate. Since metals are high in modulus, a laminate made with metal foil bonded to the surfaces of a polymer film tends to have high bending stiffness, and high stiffness is undesirable in applications requiring flexible films. Also, this laminate is subject to plastic deformation and acts much like a metal foil.

When a laminate is made with a metal foil core and faces of a polymer film such as PET, the resultant film is relatively flexible since the higher modulus material forms the center layer of the composite. The composite film is less subject to plastic deformation than the pure metal foil. Laminated films of this kind are described in this report.

Experimental procedures

Two-ply laminated poly(ethylene terephthalate) films were produced from 36- μm (1.42-mil) du Pont Mylar [12] Type A film, and three-ply laminates were produced from 23- μm (0.92-mil) du Pont Mylar Type A. The Mylar Type A films were biaxially oriented during the manufacturing process. The adhesive used for the laminates described in this report was 13- μm (0.5-mil) Sheldahl GT-100 thermoplastic polyester film (Sheldahl Corporation, Northfield, MN). A sprayed epoxy adhesive was also tried. PET film laminates were made by pressing between polished plates in a platen press. Films were bonded at 149°C under 7×10^5 Pa (100 psi) pressure for one to two minutes. The lami-

nates were then cooled under pressure. Two-ply laminates were 84 μm (3.3 mil) thick and three-ply laminates were 94 μm (3.7 mil) thick. Entrapped air at the bond lines can present a problem but can be almost eliminated by using careful fabrication techniques.

Two fiber-reinforced PET film laminates were included in this study. A two-ply laminate of a nonwoven poly(ethylene terephthalate) fabric bonded to a PET film with a thermosetting polyester adhesive was obtained from the Sheldahl Corporation. Total thickness of this laminate was 76 μm (3 mil). A three-ply laminate with two plies of 9 μm (0.35 mil) PET film and one ply of a plain weave PET fabric (85 by 85 count) bonded together with a thermosetting polyester adhesive was also obtained from the Sheldahl Corporation. The thickness of this laminate was 114 μm (4.5 mil).

Three-ply laminates with PET film faces bonded to metal foil cores were made using GT-100 thermoplastic polyester adhesive and the same press bonding operation described above. Two different metal foil cores were used in this study: a 13- μm (0.5-mil) type 302 stainless steel foil and a 25- μm (1-mil) foil of a low thermal expansion alloy (29% nickel, 17% cobalt, 53.5% iron plus trace elements). The foils were supplied in the fully annealed condition by Teledyne Rodney Metals, New Bedford, MA, and the PET film faces were 23- μm (0.92-mil) du Pont Mylar.

Yield stress, ultimate strength, ultimate elongation, and Young's modulus of the films were measured using die-cut dogbone-shaped specimens (ASTM D638, type IV). Specimen thickness was measured with a mechanical comparator to ± 0.25 μm (± 10 $\mu\text{in.}$). Tensile tests were conducted using an Instron testing machine with a 2.5-cm (1-in.) extensometer for measurement of strain in the specimens. A preload of about 7 MPa (1000 psi) was required to straighten the specimens before the extensometer was applied. The strain rates used were 0.05 min^{-1} for measurement of Young's modulus and 0.5 min^{-1} for measurement of yield and ultimate properties. At least three specimens were tested at each direction. All of these tests were conducted at approximately 23°C.

Coefficients of thermal expansion of media were determined with a du Pont 942 thermomechanical analyzer by the technique of Barrall and Logan [5]. At least three measurements were made along each direction. The specimens were 3 mm (0.118 in.) wide with 7-mm (0.275-in.) lengths between the chucks. Tests were run at a heating rate of 5°C per minute.

Coefficients of hygroscopic expansion were measured on specimens 20 cm (8 in.) square. Three specimens of

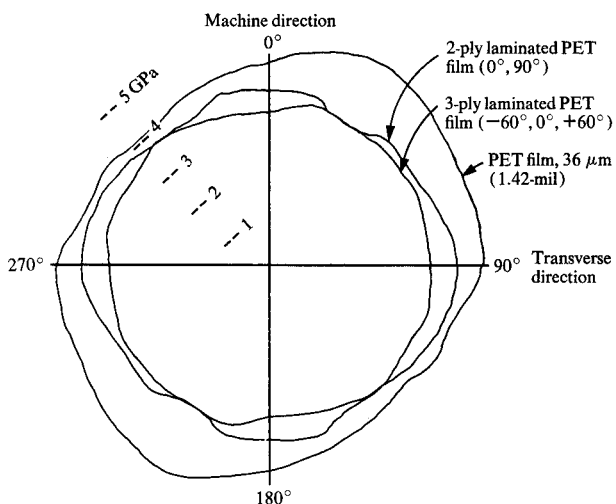


Figure 1 Young's modulus versus direction, PET film and laminated PET films.

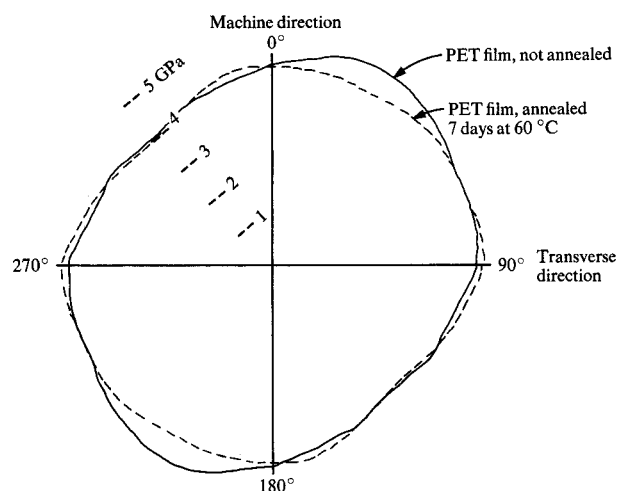


Figure 3 Young's modulus versus direction, annealed and unannealed 36- μ m (1.42-mil) PET films.

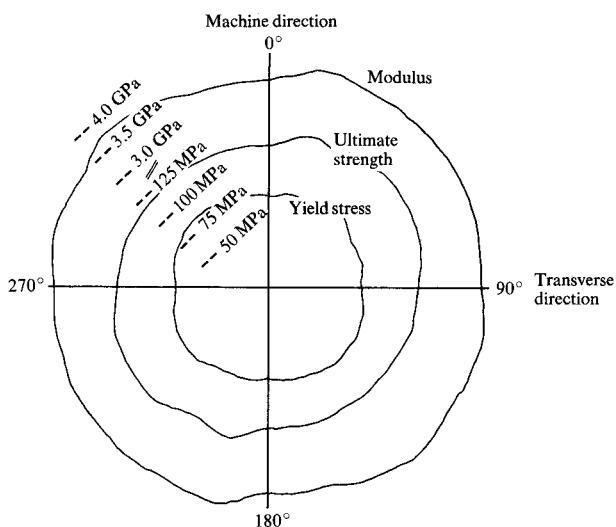


Figure 2 Mechanical properties versus direction, three-ply PET film laminate.

each material were used. Targets were made by applying patches of ink to the specimens and lightly scribing crosses into the ink. Dimensional changes were measured with a coordinate measuring microscope which reads to $\pm 0.25 \mu\text{m}$ ($\pm 10 \mu\text{in.}$). Specimens were conditioned in an environmental chamber to $8 \pm 1\%$ relative humidity at $22 \pm 1^\circ\text{C}$ for at least 24 hours.

When the chamber was opened, the specimens were pressed between glass plates, and the distances between targets were measured. The specimens were then conditioned to $80 \pm 1\%$ relative humidity at $22 \pm 1^\circ\text{C}$ for at least 24 hours. After conditioning, the specimens were

pressed between glass plates and were remeasured. The specimens were then returned to 8% relative humidity and measured again. Coefficients of hygroscopic expansion were calculated from the average of the two measurements made by increasing and then by decreasing the relative humidity.

Tests of long-term dimensional stability were conducted using the 20-cm (8-in)-square specimens with scribed targets. The specimens were conditioned to $22 \pm 1^\circ\text{C}$ and $50 \pm 1\%$ relative humidity before measurement of dimensions and were then pressed between glass plates during measurement. After the initial measurements, the specimens were placed on aluminum plates separated by spacers and put into a circulating-air oven at $60 \pm 1^\circ\text{C}$. Periodically, the specimens were removed from the oven, returned to the equilibrium temperature and relative humidity and then measured. Tests were run in triplicate.

Surface topography of the films was examined using three techniques. Profiles of the surfaces of single-ply and laminated films were obtained with a profilometer. Specimens of the films were coated with a thin gold-palladium coating for microscopic examination and were examined with a scanning electron microscope. The films were then examined by polarization interferometry using an optical microscope. Taken together, the three methods of observation can lead to a good understanding of the relative smoothness of films.

Results and discussion

Various properties of the laminated films were determined and compared to the properties of single-ply films.

Table 1 Young's modulus of PET films and film laminates.

Film	Young's modulus [GPa (10^5 psi)]			
	0 degrees ^a	45 degrees	90 degrees	135 degrees
Mylar A, 23 μ m (0.92 mil)	4.61 (6.68)	5.42 (7.86)	4.46 (6.47)	3.54 (5.13)
Mylar A, 36 μ m (1.42 mil)	4.73 (6.86)	4.96 (7.20)	4.51 (6.54)	4.01 (5.81)
Two-ply (0 degrees, 90 degrees) laminate	3.92 (5.68)	3.84 (5.57)	4.18 (6.07)	3.74 (5.42)
Three-ply (-60, 0, +60 degrees) laminate	3.43 (4.97)	3.66 (5.31)	3.56 (5.17)	3.72 (5.39)

^aOrientation with respect to machine direction of film or of one ply in laminate.

Variations in mechanical properties and coefficients of expansion with film orientation were measured. Some work was done on long-term dimensional stability of laminated films. Also, film surface roughness was examined using two microscopic techniques and a profilometer.

The two- or three-ply laminates are not symmetric in stacking sequence, and therefore the question arises as to whether they will change in flatness when the ambient temperature changes. The laminates were cooled under pressure after bonding and were quite flat when they were removed from the press. Several 30-cm-square, three-ply PET film laminates were then exposed to temperatures of first 4°C and then 52°C. Changes in flatness were measured at the edges with a feeler gauge. The flatness of the laminates changed 0.25 mm (0.010 in.) or less in this temperature range. In these flexible laminates, the asymmetric stacking sequence does not appear to cause significant warpage.

Mechanical properties

Values for Young's modulus of two PET films and two PET film laminates are given in Table 1. The single-ply films showed significant anisotropy, especially in the case of the 23- μ m (0.92-mil) film. The two-ply laminate was less anisotropic in modulus and the three-ply film had the least anisotropy. Figure 1 is a plot using polar coordinates of modulus versus direction for single-, two-, and three-ply films. It is apparent that as the number of plies in a film increased, the modulus became more nearly isotropic. Also, as the number of plies increased, the magnitude of the modulus decreased since the modulus of the adhesive is much lower than the modulus of the oriented film. The other mechanical properties of the laminates were also lower than the properties of the oriented PET films due to presence of the adhesive. In the PET film laminates, the volume fraction of adhesive was 0.22 in the two-ply film and 0.27 in the three-ply material.

Figure 2 shows the mechanical properties of a three-ply PET film laminate with the plies oriented at -60, 0, and +60 degrees. Modulus values ranged from 3.43 GPa to 3.72 GPa (4.97×10^5 psi to 5.39×10^5 psi) and varied from the median value of 3.58 GPa (5.19×10^5 psi) by less than 5%. Yield stress varied from 76.5 MPa to 86.2 MPa (11.1×10^3 psi to 12.5×10^3 psi) in the three-ply laminate, a 6% variation from the mean of 81.4 MPa. The ultimate strength values ranged from 118 MPa to 134 MPa (17.1×10^3 psi to 19.5×10^3 psi) and varied from the mean strength of 18.3 MPa by 7%. The ultimate elongation ranged from 33 to 46% in the laminate and the variation from the mean value for elongation was 16%. In general, it can be stated that the three-ply PET film was nearly isotropic in mechanical properties.

Annealing a PET film may lead to a reduction in anisotropy by allowing stresses in noncrystalline regions of the film to relax. Figure 3 is a polar plot of the Young's modulus of 36- μ m (1.42-mil) PET film before and after annealing. Annealing the film at 60°C, which is below the glass transition temperature of the PET, caused no reduction in the modulus of the film but did make the material somewhat less anisotropic. However, the annealed film was still much more anisotropic than the laminated PET films.

The mechanical properties of composite laminated films are given in Table 2. The laminates reinforced with nonwoven and woven PET fabrics showed considerable anisotropy in strength and elastic modulus. In the film with the nonwoven fabric reinforcement, modulus values ranged from 2.05 GPa to 3.11 GPa (2.98×10^5 psi to 4.51×10^5 psi) and strength values ranged from 65.5 MPa to 104.1 MPa (9.5×10^3 psi to 15.1×10^3 psi). The film reinforced with the woven PET fabric showed higher strength and stiffness at 0° and 90° directions and lower values at 45° and 135°, as would be expected. Modulus

Table 2 Mechanical properties of composite film laminates.

<i>Reinforcement</i>	<i>Direction</i>	<i>Young's modulus</i> [GPa (10 ⁵ psi)]	<i>Yield stress</i> [MPa (10 ³ psi)]	<i>Ultimate strength</i> [MPa (10 ³ psi)]	<i>Ultimate elongation (%)</i>
Nonwoven PET fabric	0 degrees	2.52 (3.65)	50.3 (7.3)	88.9 (12.9)	34
	45 degrees	2.05 (2.98)	51.0 (7.4)	65.5 (9.5)	58
	90 degrees	2.44 (3.54)	51.7 (7.5)	66.9 (9.7)	53
	135 degrees	3.11 (4.51)	53.8 (7.8)	104.1 (15.1)	34
Woven PET fabric	0 degrees	1.14 (1.66)	no yield pt	54.5 (7.9)	31
	45 degrees	0.57 (0.82)	16.5 (2.4)	28.3 (4.1)	44
	90 degrees	0.74 (1.07)	no yield pt	54.5 (7.9)	34
	135 degrees	0.52 (0.75)	15.9 (2.3)	23.4 (3.4)	48
Stainless steel foil	0 degrees	15.7 (22.8)	85 (12.3) ^a	128 (18.6)	25
	45 degrees	19.5 (28.3)	119 (17.3)	143 (20.7)	22
	90 degrees	18.8 (27.3)	113 (16.4)	141 (20.4)	21
	135 degrees	18.8 (27.2)	110 (15.9)	190 (27.6)	25
Low expansion alloy foil	0 degrees	32.1 (46.6)	136 (19.7)	174 (25.3)	24
	45 degrees	33.2 (48.2)	141 (20.5)	206 (29.9)	30
	90 degrees	33.9 (49.2)	148 (21.4)	191 (27.7)	24
	135 degrees	32.8 (47.6)	137 (19.8)	172 (24.9)	26

^aYield stress for metal foil core laminates measured as 2% offset yield.**Table 3** Coefficient of linear thermal expansion of PET film and laminates at 25 to 50°C.

<i>Film</i>	<i>Coefficient of linear thermal expansion [10⁻⁵(C°)⁻¹]</i>			
	<i>0 degrees^a</i>	<i>45 degrees</i>	<i>90 degrees</i>	<i>135 degrees</i>
Mylar A, 23 μm (0.92 mil)	1.40	0.96	1.26	2.01
Mylar A, 36 μm (1.42 mil)	1.62	0.95	1.00	1.73
Two-ply (0 degrees, 90 degrees) PET film laminate	1.55	1.42	1.86	1.70
Three-ply (-60, 0, +60 degrees) PET film laminate	2.00	2.05	2.15	2.08
Nonwoven PET fabric laminate	2.20	2.31	1.98	1.03
Woven PET fabric laminate	1.76	1.87	—	—
Stainless steel foil laminate	2.12	—	2.12	—
Low expansion alloy foil laminate	0.86	0.84	0.86	0.89

^aOrientation with respect to machine direction of film or of one ply in laminate.**Table 4** Coefficient of hygroscopic expansion of PET films and laminates, 8 to 80% relative humidity, 23°C.

<i>Film</i>	<i>Coefficient of hygroscopic expansion [10⁻⁶ (%RH)⁻¹]</i>			
	<i>0 degrees^a</i>	<i>45 degrees</i>	<i>90 degrees</i>	<i>135 degrees</i>
Mylar A, 23 μm (0.92 mil)	6.2	5.4	6.1	9.6
Mylar A, 36 μm (1.42 mil)	7.3	6.6	7.1	7.7
Three-ply (-60, 0, +60 degrees) PET film laminate	6.3	6.0	6.4	6.3
Woven PET fabric laminate	5.8	5.8	6.0	5.6
Low expansion alloy foil laminate	1.2	1.7	1.3	1.2

^aOrientation with respect to machine direction of film or of one ply in laminate.

Table 5 Shrinkage due to stress relaxation of PET films and laminates, 1344 hours at 60°C.

Film	Shrinkage (%)			
	0 degrees ^a	45 degrees	90 degrees	135 degrees
Mylar A, 23 μm (0.92 mil), not annealed	0.113	—	0.100	—
Mylar A, 36 μm (1.42 mil), not annealed	0.066	0.059	0.052	0.067
annealed	0.013	0.012	0.018	0.013
Two-ply PET film laminate, not annealed	0.050	0.052	0.060	0.057
Three-ply PET film laminate, not annealed	0.048	0.060	0.053	0.068
annealed	0.014	0.004	0.008	0.015
Low expansion alloy reinforced laminate, not annealed	0.019	0.003	0.018	0.017

^aOrientation with respect to machine direction of film or of one ply in laminate.

values ranged from 0.52 GPa to 1.14 GPa (0.75×10^5 psi to 1.66×10^5 psi) and strength values ranged from 23.4 MPa to 54.5 MPa (3.4×10^3 psi to 7.9×10^3 psi).

Laminated films with metal foil cores were more more nearly isotropic than the films reinforced with PET fabrics. Because of the high modulus of the foils, the foil-reinforced laminates were much stiffer than the other films studied. In the stainless steel foil-reinforced laminate, modulus values ranged from 15.7 GPa to 19.5 GPa (2.28×10^6 psi to 2.83×10^6 psi) and strength values ranged from 128 MPa to 190 MPa (18.6×10^3 psi to 27.6×10^3 psi). The film reinforced with the low expansion alloy film was essentially isotropic in elastic modulus, with values ranging from 32.1 GPa to 33.9 GPa (4.66×10^6 psi to 4.92×10^6 psi). In strength, this film showed greater anisotropy, with values ranging from 172 MPa to 206 MPa (24.9×10^3 psi to 29.9×10^3 psi).

Coefficient of linear thermal expansion

Coefficients of linear thermal expansion of PET films and laminated films are given in Table 3. The single-ply PET films and the laminate with the nonwoven PET fabric reinforcement showed pronounced anisotropy in thermal expansion coefficients, with a factor of two difference between high and low values in a given film. The PET film laminates were significantly more uniform in coefficient of thermal expansion than the single-ply films, and the three-ply film was nearly isotropic. The PET film laminates have coefficients of expansion which are higher than the coefficients for the single-ply material. This is to be expected since the coefficient of thermal expansion of the adhesive is several times greater than the coefficient for oriented PET film.

The laminate reinforced with the low expansion alloy foil had the lowest values for coefficient of linear thermal expansion of all of the films tested, as would be expected. In addition, this film is essentially isotropic in in-plane thermal expansion.

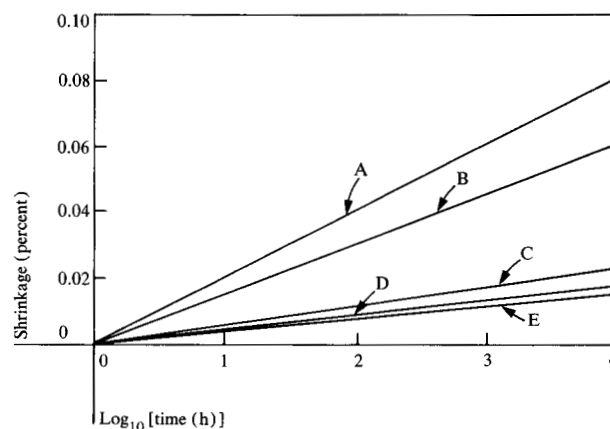


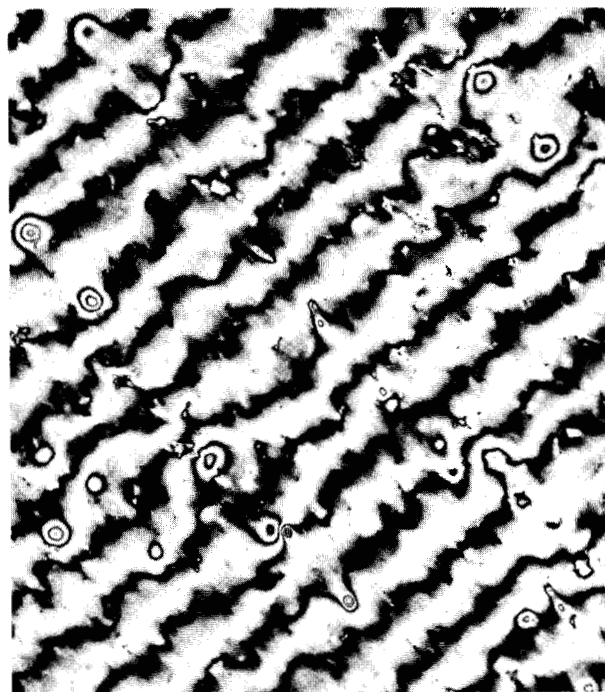
Figure 4 Long-term dimensional stability of films and laminates at 60°C (machine direction of single-ply film and machine direction of one ply in three-ply laminate): curve A, PET film, 36 μm , not annealed; curve B, 3-ply PET film laminate, not annealed; curve C, low expansion foil laminate, not annealed; curve D, 3-ply PET film laminate, annealed; curve E, PET film, 36 μm , annealed.

Coefficient of hygroscopic expansion

Table 4 shows coefficients of hygroscopic expansion of single-ply PET films and laminated films. The coefficient of hygroscopic expansion was anisotropic in the single-ply films, especially in the thinner film. In the three-ply PET film laminate, the variation in coefficient of hygroscopic expansion with direction was much less. The laminate reinforced with woven PET fabric has slightly smaller coefficients of hygroscopic expansion than the three-ply laminate and is also nearly isotropic in this property. Because the reinforcing metal foil absorbs virtually no water, the laminate made with the low expansion alloy had very small coefficients of hygroscopic expansion. This laminate showed some anisotropy in hygroscopic expansion but the absolute variation in coefficient was still quite small.

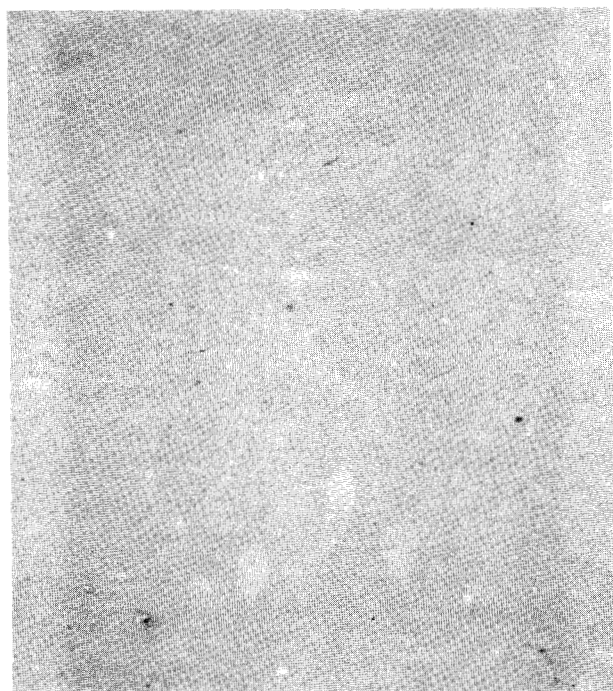


(a)



(b)

Figure 5 Photomicrographs of 23- μm (0.92-mil) PET film: (a) by scanning electron microscopy, 400 $\times$; (b) by polarization interferometry, 360 $\times$.



(a)



(b)

Figure 6 Photomicrographs of three-ply laminated PET film: (a) by scanning electron microscopy, 400 $\times$; (b) by polarization interferometry, 360 $\times$.

Long-term dimensional stability

An oriented polymer film is subject to stress relaxation which results in irreversible in-plane shrinkage of the film. The magnitude of the shrinkage observed depends on the time-temperature-stress history of the film. In this study, dimensional stability tests were conducted by exposing specimens to dry air at 60°C. Specimens were brought to equilibrium at 23°C and 50% relative humidity before each measurement of dimensions. Single-ply films were annealed by placing them on aluminum plates in an oven at 107°C for two hours. The annealed PET film laminates were made by first annealing single sheets of film and then bonding them.

At 60°C, all of the films showed some shrinkage. As would be expected, shrinkage at a given temperature is rapid at first and then occurs more slowly. Values for shrinkage of single-ply and laminated films after 1344 hours at 60°C are given in Table 5. The thinner PET film had the most shrinkage, presumably due to higher induced stresses during manufacture of the film. Annealing significantly reduces shrinkage of both single-ply and laminated films. The unannealed PET film laminates had about the same shrinkage as the 36- μm single-ply film. Because the plies cannot shrink freely during the bonding operation, the lamination process does not allow stress relaxation to take place. A separate annealing process, either before or after lamination, appears necessary if laminated PET films are to have good long-term dimensional stability.

The laminate reinforced with the low expansion alloy foil had low values for shrinkage at 60°C, about the same as for the annealed PET films. Annealing this composite film could result in even smaller values for shrinkage.

Figure 4 shows the dependence of shrinkage on time at 60°C for the experimental films. The plots of shrinkage against $\log_{10}$ time are essentially linear over the time period included in this study.

All of the films were anisotropic with respect to shrinkage. However, as with other properties, when the shrinkage values are low, the absolute variation in shrinkage with direction is quite small.

Surface topography

Many applications for flexible films require that the films be as smooth as possible. Because asperities in PET films are relatively small, the techniques used to detect them require relatively high magnifications and therefore each observation is of a small surface area. A large number of observations are needed to characterize the topography of films. However, if two films are significantly different

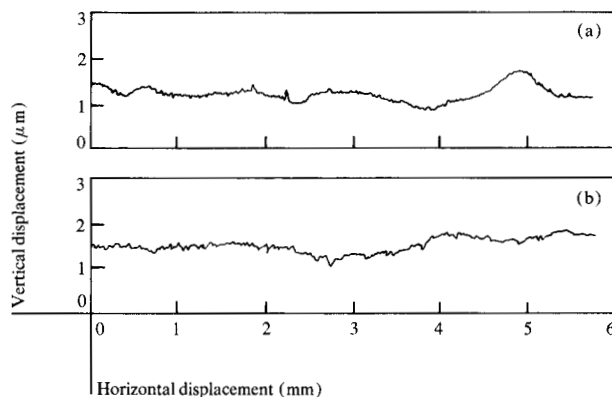


Figure 7 Surface profiles: (a) 23- μm (0.92-mil) PET film; (b) three-ply laminated PET film.

in surface topography, a few observations may be sufficient to make general comparisons between the films.

Figure 5 shows a 23- μm (0.92-mil) PET film as observed by scanning electron microscopy and polarization interferometry. Figure 6 shows corresponding photomicrographs of a three-ply laminated PET film. Typical profile traces of single-ply and three-ply films are presented in Fig. 7. It is apparent that the larger asperities in the single-ply film are reduced in height when the film is laminated between smooth surfaces. The larger asperities are simply pressed into the surface during the bonding operation. However, laminated films contain many small asperities formed by defects in the surfaces used for bonding and by dust particles which are pressed into the material.

Conclusions

Laminated poly(ethylene terephthalate) films can be produced with nearly isotropic mechanical properties and coefficients of thermal and hygroscopic expansion. Because the adhesive is lower in modulus and higher in thermal expansion than oriented PET film, laminated films have lower modulus and higher coefficients of thermal expansion than single-ply PET films. Both single-ply and laminated PET films are subject to shrinkage over long periods of time. Annealing significantly reduces the shrinkage of these films when they are subjected to a temperature of 60°C. When polished plates or rolls are used to laminate the films, the larger asperities are reduced in height by pressing the asperities into the surface.

Of the reinforced films included in this study, the film reinforced with the low expansion alloy foil had the best dimensional stability and also was the most nearly isotropic. However, this film was much stiffer than the poly-

mer films. Because the composite films offer the possibility of superior dimensional stability, further work on these films is warranted.

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